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Hammershøi, Peter S.; Janssens, Ton V.W.; Jensen, Anker D.

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Lifetime impact of SO_2-poisoning of a Cu-CHA catalyst for NH_3-SCR

Peter S. Hammershøi[a,b], Ton V. W. Janssens[b], Anker D. Jensen[a,*]

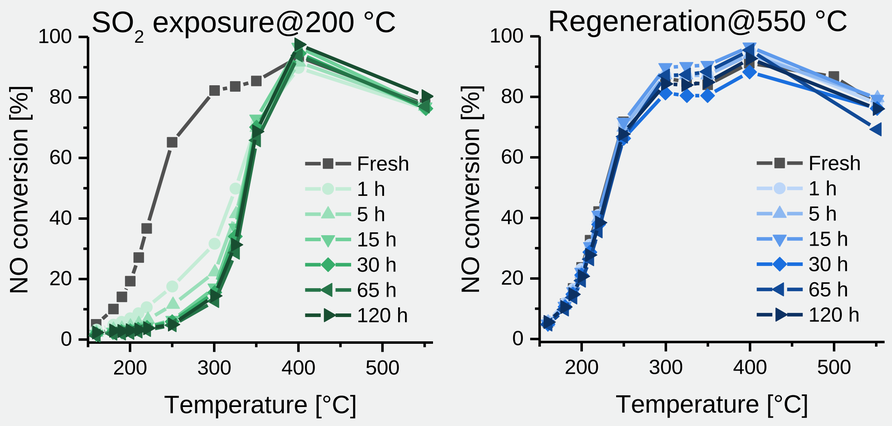
[a] *Department of Chemical Engineering, Technical University of Denmark, Søltofts Plads B229, 2800 Kgs. Lyngby, Denmark*

[b] *Umicore Denmark ApS, Nøjsomhedsvej 20, 2800 Kgs. Lyngby, Denmark*

** Corresponding author. Phone: +45 45252841, e-mail: aj@dtu.kt.dk*

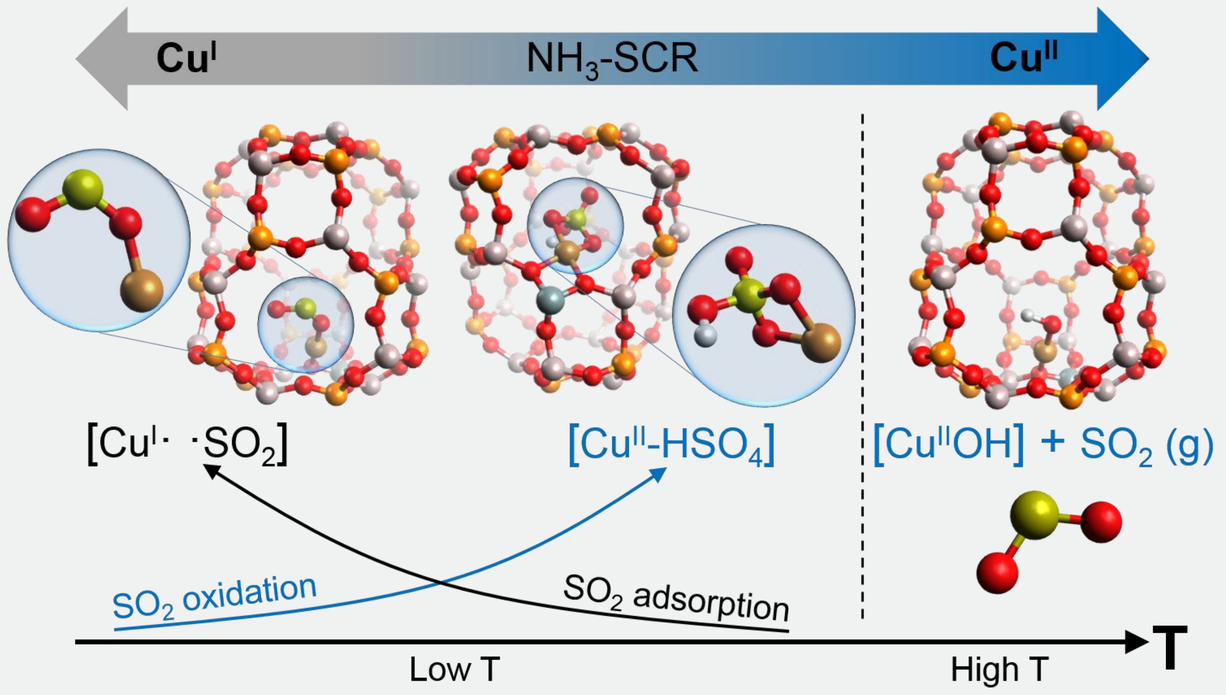
Background

The highest activity for removal of NO_x (NO and NO_2) in the temperature range 180-250 °C, by selective catalytic reduction (SCR) with NH_3 in the after treatment system of heavy-duty vehicles, is obtained with Cu-zeolite catalysts. Improved fuel efficiency that is necessary for lower CO_2 emissions, has led to colder exhausts, which has increased the importance of efficient NO_x removal in that temperature range. However, Cu-zeolites deactivate by the inevitable presence of SO_2 in diesel exhaust, and therefore, we have investigated if the lifetime impact of SO_2 on the activity of a Cu-CHA zeolite catalyst is prohibitive for practical application.



Typical SO_2-poisoning behavior

- Exposure to SO_2 results in significantly lower NO conversions in the temperature range 180-350 °C
- Regeneration at 550 °C in SO_2-free gas can restore the predominant part of the drop in NO conversion inferred by SO_2

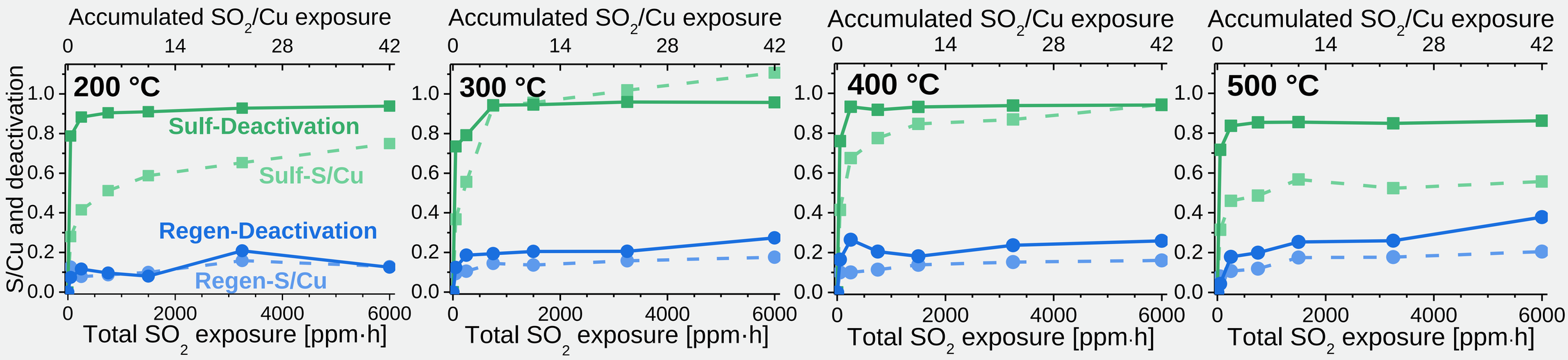


Potential lifetime SO_2/Cu exposure

Assumptions for the lifetime of a heavy-duty vehicle:

- Distance: 800 000 km
- Average mileage: 30 L diesel/ 100 km
- Diesel: 5 wt ppm S
  (Ultra-low sulfur diesel in: EU: <10 wt ppm S, USA: <15 wt ppm S)
- Cu-CHA: 1.5 kg with 3 wt% Cu (45 g Cu)

- Potential accumulated lifetime SO_2/Cu exposure: **44**



Experimental

Catalyst: Cu-CHA powder with Si/Al of 14.6 and Cu/Al of 0.42 (2.76 wt% Cu on dry matter basis), and of sieve fraction 150-300 µm.
SCR activity measurements: 5 mg catalyst, 500 ppmv NO, 533 ppmv NH_3, 10 % O_2, 5 % H_2O, with N_2 at a total flow rate of 225 NmL/min.
SO_2 exposure: 50 ppmv SO_2, 10 % O_2 and 5 % H_2O with N_2 to 1.67 NL/min at 200, 300, 400 or 500 °C for 1, 5, 15, 30, 65 or 120 h.
Regeneration: 6 h at 550 °C in 10 % O_2 and 5 % H_2O gas.
ICP-OES: S/Cu ratios of the sulfated and regenerated catalysts.

Conclusions

- Deactivation occurs fast regardless of exposure temperature
  - Maximum deactivation levels reached already at low S/Cu ratios
- Regeneration at 550 °C can limit deactivation to 20 % throughout the catalyst lifetime
- Practical application of Cu-CHA as SCR catalyst is contingent on:
  - Efficient regeneration
  - Use of ultra-low sulfur diesel

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